Solution Phase Photochemistry in Run 21
LCLS Virtual Town Hall

Thomas Wolf, LCLS Chemical Sciences Department head
Roberto Alonso-Mori, Bio-Chemistry Group lead
03/21/2022

U.S. DEPARTMENT OF ENERGY
Office of Science

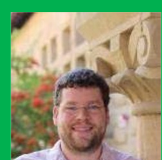
SLAC NATIONAL ACCELERATOR LABORATORY

Outline

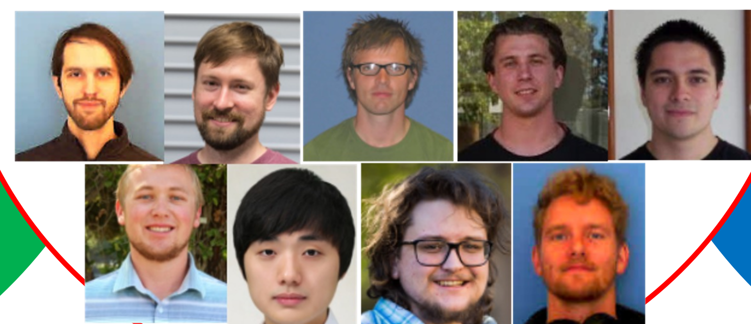
- Solution Phase Photochemistry
  - LCLS Team
  - Early Science Soft X-rays at High Rep Rate
  - Hard X-ray Capabilities
- Gas Phase Photochemistry
  - LCLS Team
  - Early Science Soft X-rays at High Rep Rate
  - Hard X-ray Capabilities

Solution Phase Photochemistry in the SRD Department Structure

Chemical Sciences
Department
Thomas Wolf

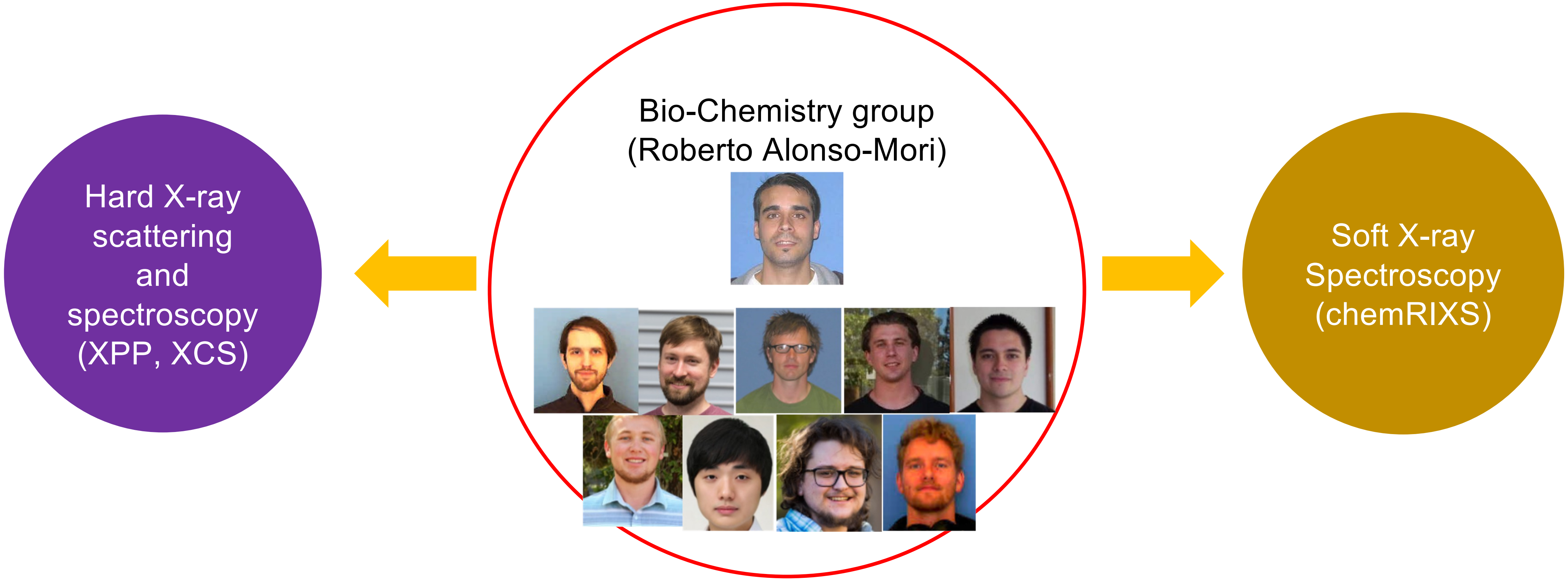


Bio-Chemistry group
(Roberto Alonso-Mori)



Biology Department
Mark Hunter

Techniques and Instruments Supported by the Group



Points of contact - by science area and by instrument

LCLS Instrument Contacts:

- **Time-resolved AMO (TMO)** - James Cryan (**jcryan@slac.stanford.edu**)
- **ChemRIXS** - Georgi Dakovski (**dakovski@slac.stanford.edu**) or Kristjan Kunnus, (**kristjan@slac.stanford.edu**)
- **qRIXS** - Georgi Dakovski (**dakovski@slac.stanford.edu**)
- **X-ray Pump Probe (XPP)** - Diling Zhu (**dlzhu@slac.stanford.edu**)
- **X-ray Correlation Spectroscopy (XCS)** - Matthieu Chollet (**mchollet@slac.stanford.edu**)
- **Macromolecular Femtosecond Crystallography (MFX)** - Alex Batyuk (**batyuk@slac.stanford.edu**)
- **Coherent X-ray Imaging (CXI)** - Meng Liang (**mliang@slac.stanford.edu**)
- **Matter in Extreme Conditions (MEC)** - Gilliss Dyer (**gilliss@slac.stanford.edu**)

LCLS Scientific Department Head Contacts:

- Atomic, Molecular and Optical Sciences - James Cryan (**jcryan@slac.stanford.edu**)
- Biological Sciences - Mark Hunter (**mhunter2@slac.stanford.edu**)
- Chemical Sciences - Thomas Wolf (**thomas.wolf@slac.stanford.edu**)
- Laser Science - Joe Robinson (**jsrob@slac.stanford.edu**)
- Materials Science - Apurva Mehta (**mehta@slac.stanford.edu**)
- Materials in Extreme Conditions - Gilliss Dyer (**gilliss@slac.stanford.edu**)

Overview: LCLS-II Upgrade for TMO and NEH2.2

SLAC

- LCLS-II superconducting accelerator will startup during 2022/23
- First Light planned for November 2022
- For Run 21, the soft X-ray instruments will focus on using the high rep-rate beam
  - TMO, ChemRIXS, qRIXS
  - Technical commissioning followed by an LCLS-led, community-wide ‘Early Science’ period
  - No PRP proposals for Run 21 for these instruments
  - Users should submit ideas for the “Early Science” experiments (see next slides)

Soft X-ray Early Science Run 21

Soft X-ray Spectroscopy (chemRIXS)

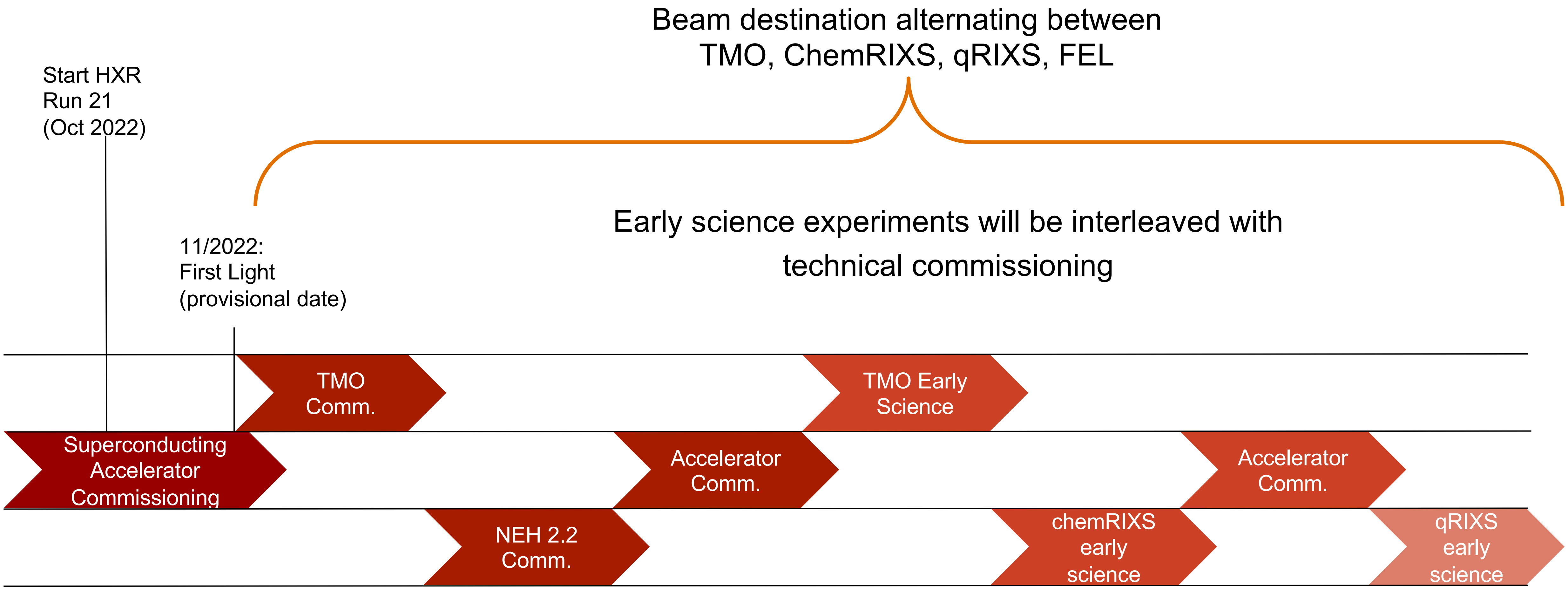
The Early Science process

- Motivation:
  - The complexity brought by high repetition rate operation warrants the implementation of a 3-step approach:
    1. Allocate sufficient time for technical commissioning of beamline and instruments at high repetition rate
    2. Early Science, bridging the gap from technical commissioning of new instrumentation to regular user access
    3. PRP proposals (planned for the next Run)
  - Enables a more flexible response to emerging LCLS-II performance, and beamline/instrument readiness
- Early Science
  - Based on ideas solicited from the community
  - Led by LCLS staff, with broad involvement from the community
  - Overseen by the LCLS Scientific Advisory Committee (SAC) and the Instrument Advisory Panels (IAPs)
- Interested groups should contact the relevant department heads - deadline 30 March
  - **TMO**: James Cryan (AMOS, jcryan@slac.stanford.edu)
  - **ChemRIXS**: Thomas Wolf (Chemical Sciences, thomas.wolf@slac.stanford.edu)
  - **qRIXS**: Apurva Mehta (Materials Sciences, mehta@slac.stanford.edu)
- Experiment ideas will then be prioritized by LCLS staff and the instrument advisory panels.
- The resultant early science plans will be advertised to the user community to solicit participation.

Timeline For Early Science Program

- **March 30, 2022**: Deadline for Letters of Interest to LCLS (same date as regular proposals)
  - One-page summary of science / instrument areas of interest, or
  - Bulleted list of experimental ideas
- **April - June 2022**: LCLS engages with User Community to develop the plan.
- **June 30, 2022:** LCLS announces Early Science experiments to User Community
- **September 1, 2022**: Deadline for interested users to submit a description of their proposed contribution to the specific Early Science experiments.
  - Experiments are open enrollment, subject to forming a balanced onsite team.
- **November 2022:** Provisional date for 'First Light' from SCRF beam, followed by:
  - FEL commissioning
  - Beamline/instrument commissioning
  - Early Science (likely in early 2023 onwards)

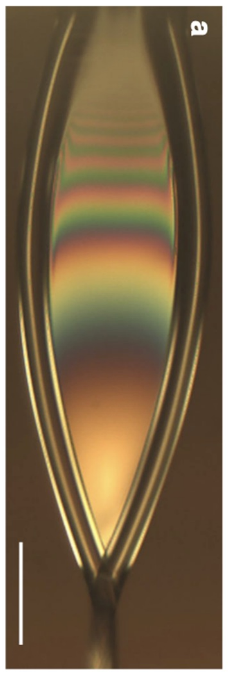
Early Science during Run 21 will follow a phased approach between the instruments, interleaved with FEL ramp-up

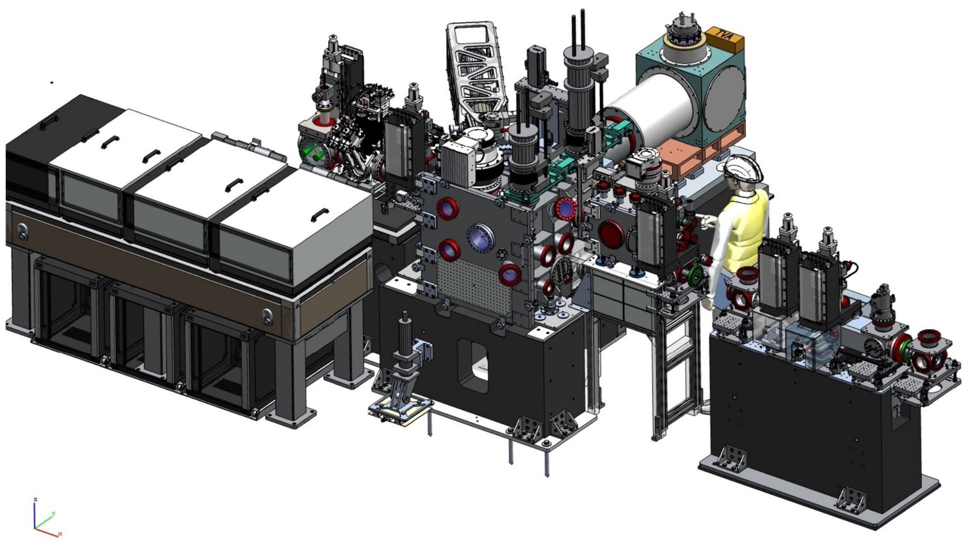


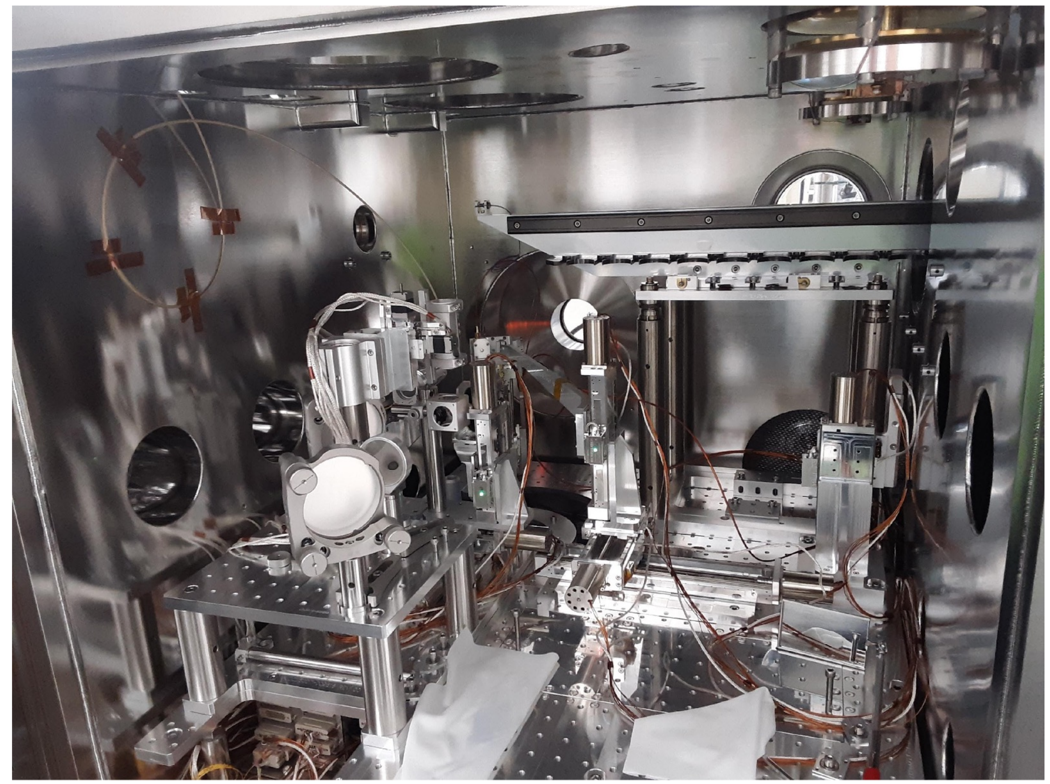
The chemRIXS Endstation

Endstation for in-vacuum liquid sheet jets
Available detectors:

- APDs for Total fluorescence yield detection
- Andor camera
- Variable line spacing spectrometer







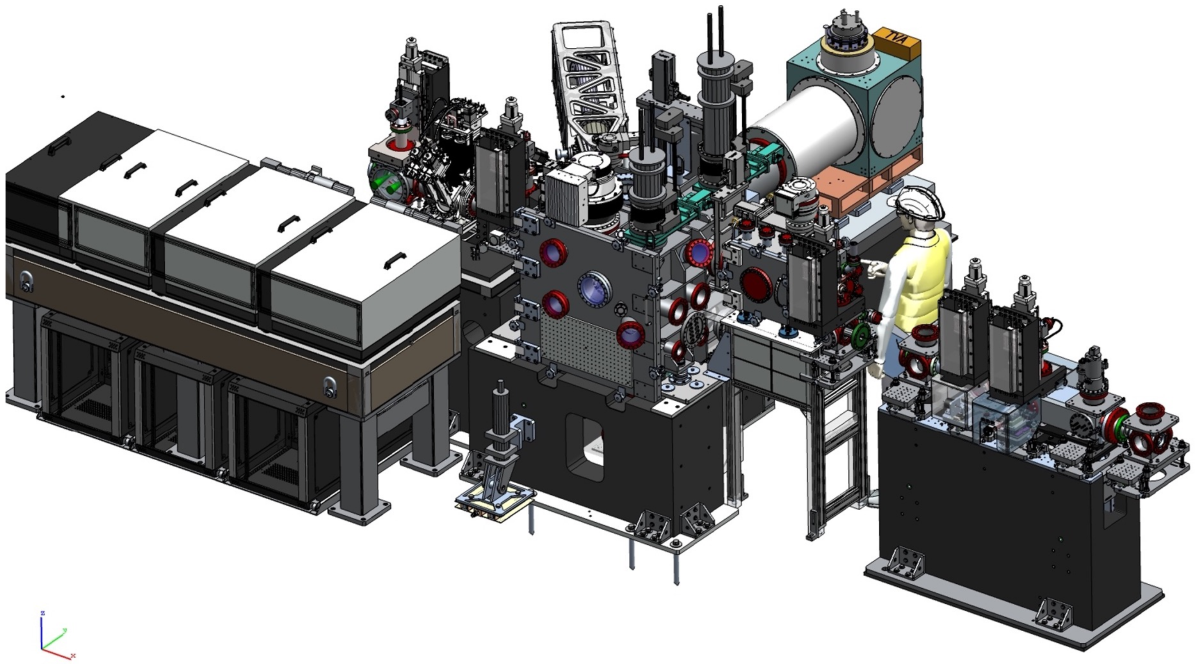
X-ray and Laser Parameters for chemRIXS in Run 21

X-ray Parameters	
Repetition rate (Hz)	Up to 50 kHz
Energy Range (eV)	250 - 1100
Pulse Duration	20 fs (nominal)
Energy per pulse (downstream of mono)	>100 nJ
Beamline resolving power	> 2000
Spot Size, FWHM (range)	10 - 1000 (um) diameter
Polarization	Linear, Horizontal

<table>
<tr><th colspan="4">Laser Parameters</th></tr>
<tr><td>Repetition rate (Hz)</td><td colspan="3">Synchronized up to 33 kHz</td></tr>
<tr><td rowspan="2">Wavelength</td><td rowspan="2">800 nm</td><td rowspan="2">400 nm</td><td>High Risk</td></tr>
<tr><td>266 nm</td></tr>
<tr><td>Pulse Duration</td><td>< 25 fs</td><td>< 50 fs</td><td>< 50 fs</td></tr>
<tr><td>Energy per pulse (on target)</td><td>300 μJ</td><td>> 30 μJ</td><td>~ 3 μJ</td></tr>
<tr><td>Spot Size, FWHM (800 nm)</td><td colspan="3">50 to 100 um</td></tr>
<tr><td>Polarization</td><td colspan="3">Variable: linear, circular</td></tr>
<tr><td>Angle</td><td colspan="3">~0.5 deg angle with x-ray beam</td></tr>
<tr><td>Arrival Time Monitor</td><td colspan="3">< 20 fs accuracy in x-ray/laser arrival time tagging.</td></tr>
</table>

User Involvement in Early Science

- Interested groups should contact Thomas (thomas.wolf@slac.stanford.edu)
- Department head collects experiment ideas and prioritize together with the chemRIXS instrument team and the instrument advisory panel.
- LCLS communicates consolidated early science plan with user community and broadly advertise participation.
- LCLS updates interested user groups on adjustments to the early science plan.



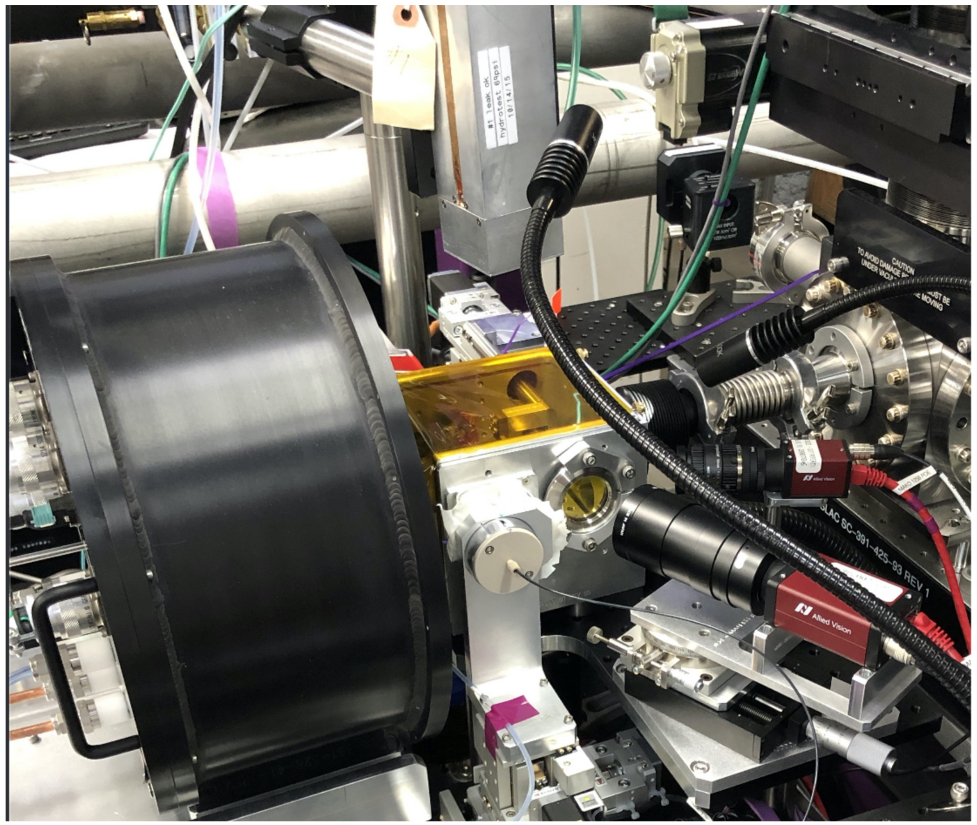
Hard X-ray Capabilities for User Proposals in Run 21

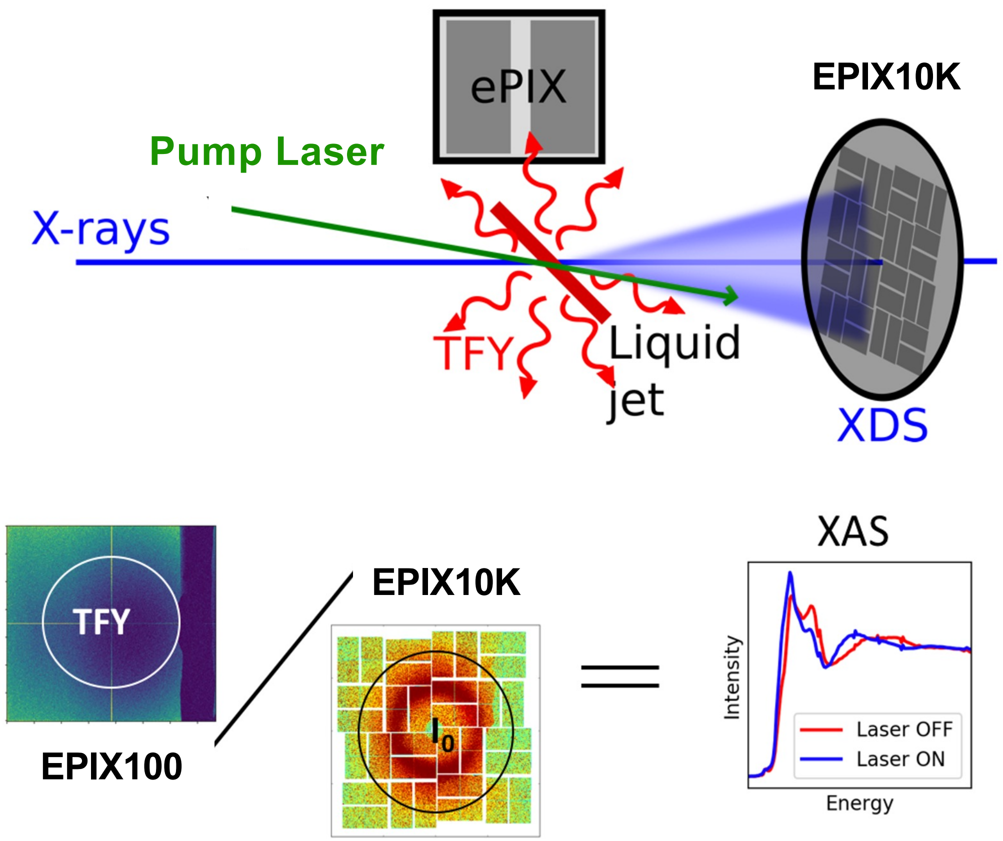
Hard X-ray scattering and spectroscopy (XPP, XCS)

XPP Standard Configuration #2: Liquid Phase XAS

Time Resolved Hard X-ray XAS

120 Hz



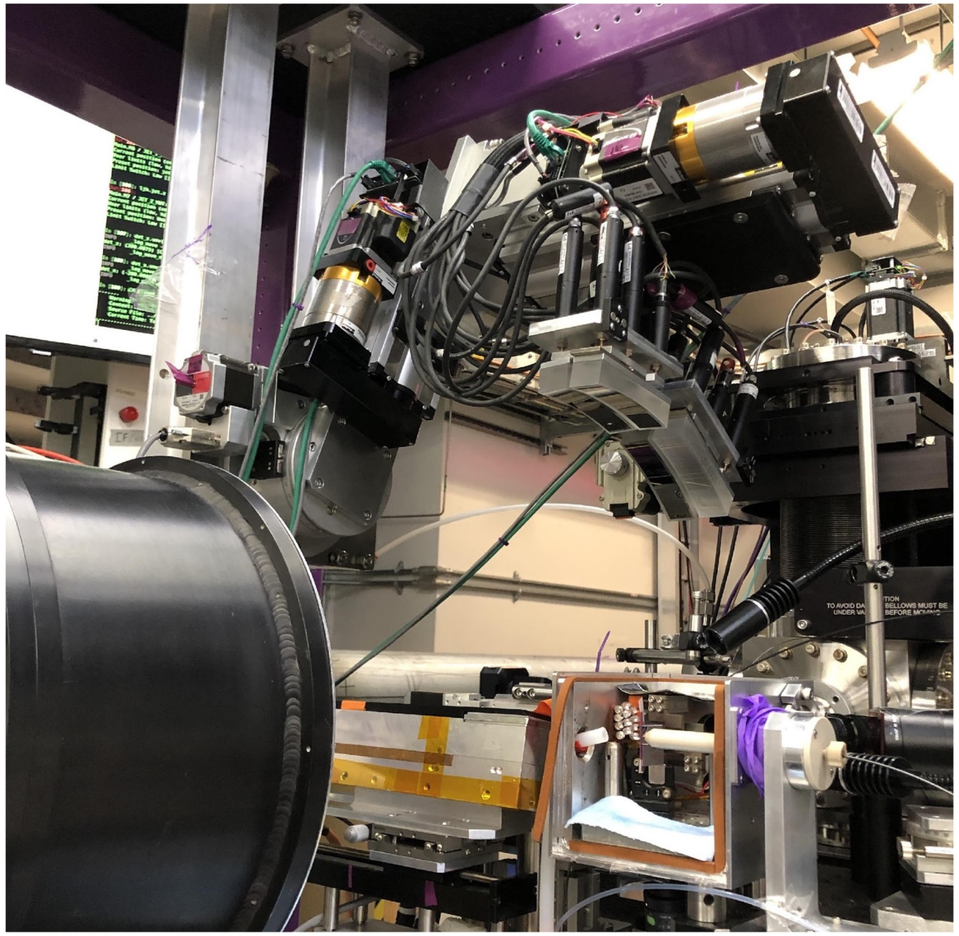


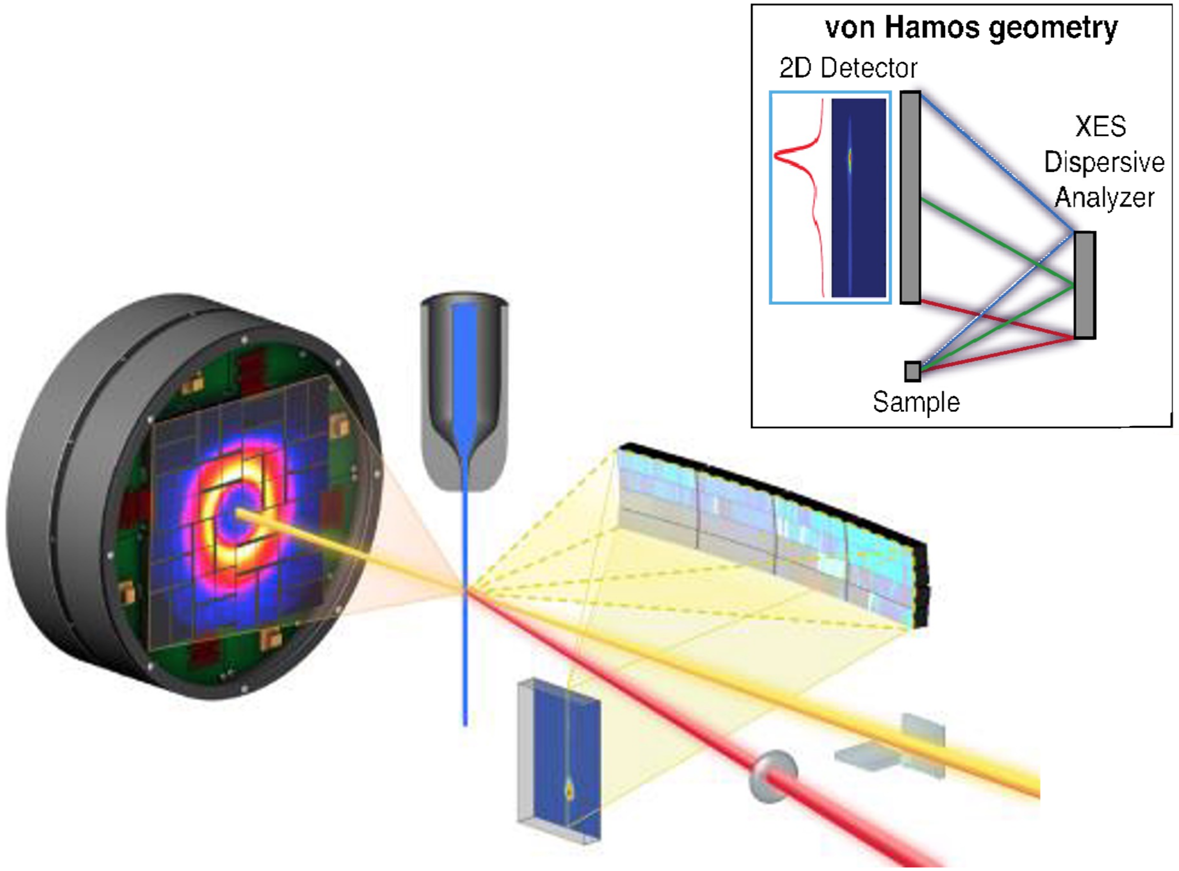
https://lcls.slac.stanford.edu/instruments/xpp/standard-configurations

XCS Standard Configuration #1: Liquid Phase XES/XDS

Time Resolved Hard X-ray XES +XDS

120 Hz

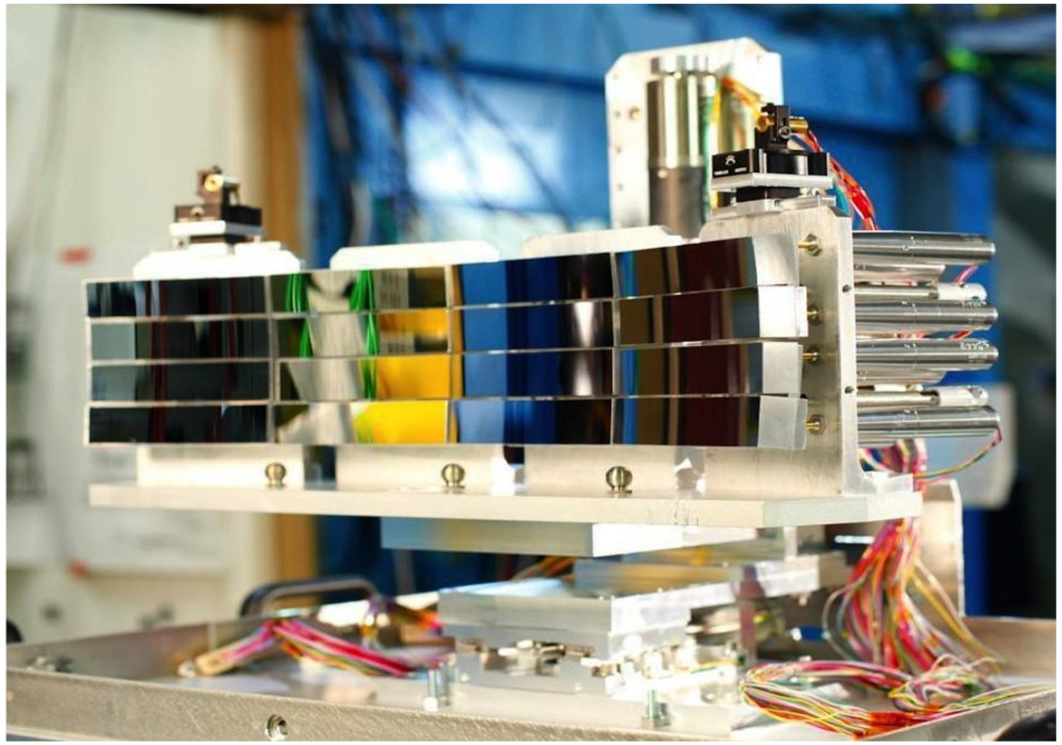




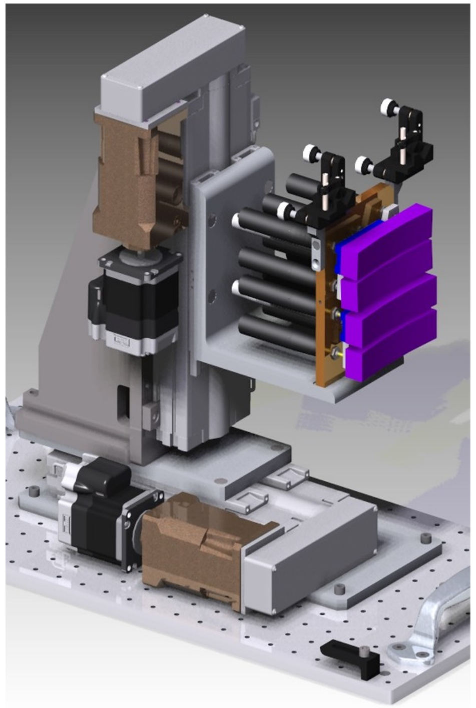
https://lcls.slac.stanford.edu/instruments/xcs/standard-configurations

Hard X-ray Spectroscopy at LCLS: XES

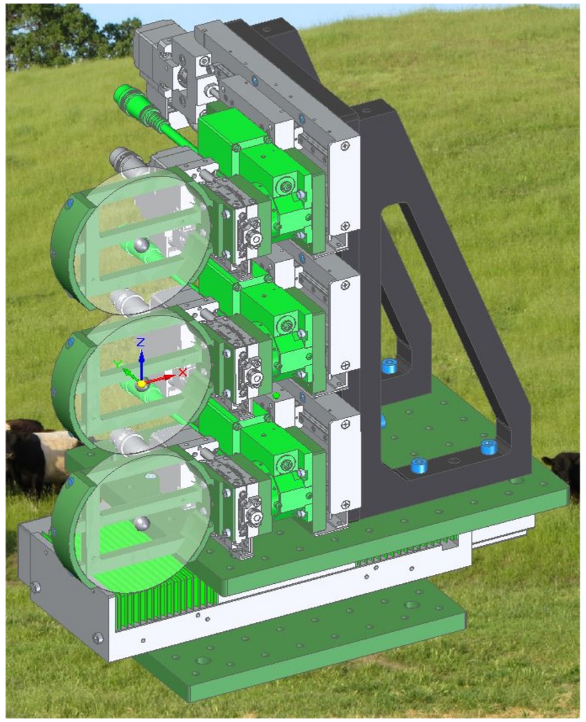
Existing LCLS multi-crystal X-ray Emission Spectrometers



16 crystal energy dispersive von Hamos



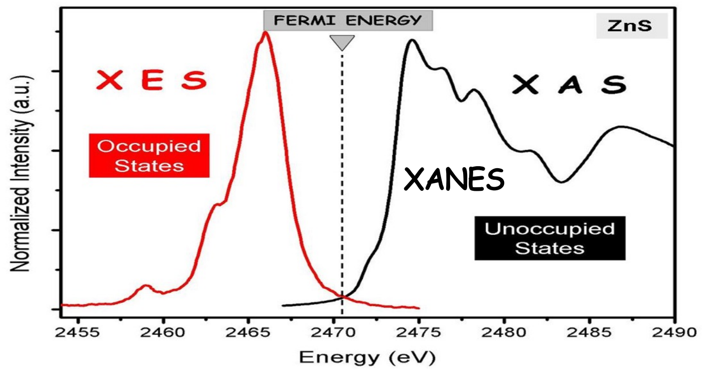
4 crystal E. dispersive von Hamos

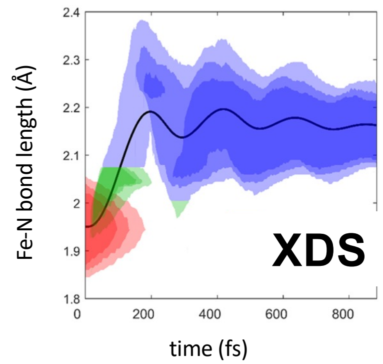


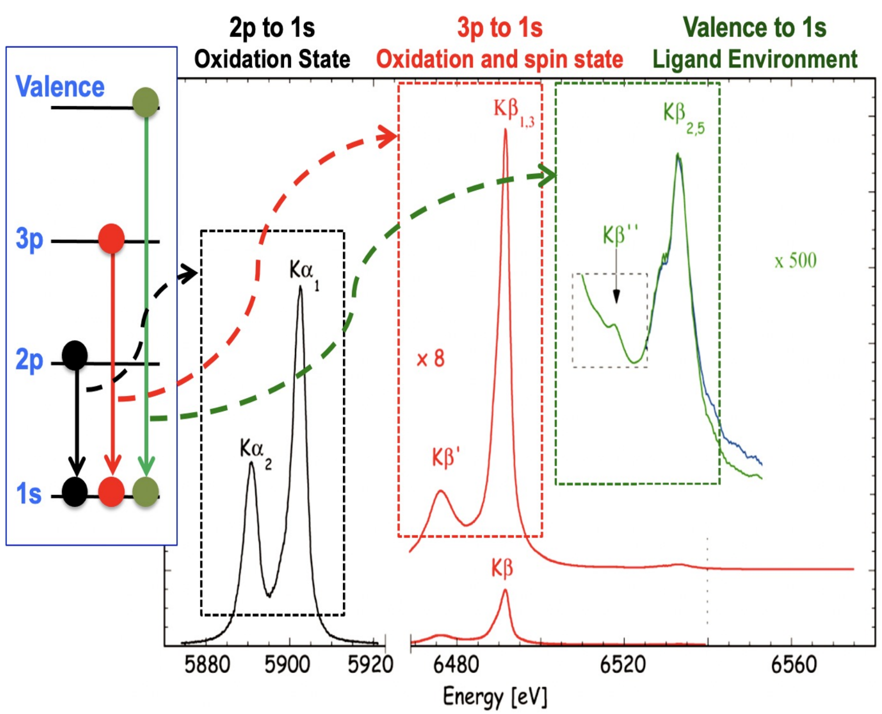
3 crystal scanning Rowland

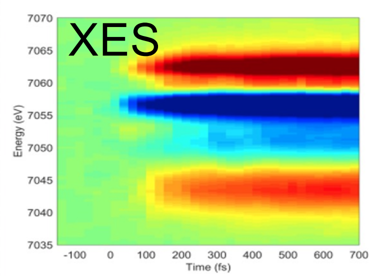
Alonso-Mori et al. RSI, 83 (2012)

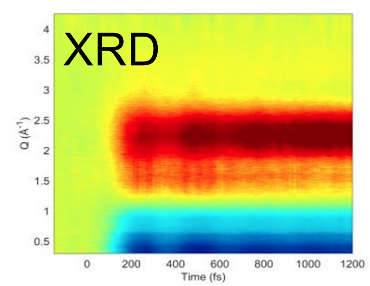
Spectroscopy/XDS Measurements:











Standard:

- XAS (XANES)
- XDS (0.2 to 5 q-range)
- HighEnergie XDS expand q (<25keV)
- Kα, $K\beta_{1,3}$ and $K\beta_{2,5}$ XES of 3d TM
- L XES of 5d TM

Not so Standard:

- Resonant XES measurements
  - RIXS (von Hamos)
  - HERFD (Rowland)
- EXAFS

Standard Configuration Parameters Table:

Sample Delivery:

Standard:

- Round jets (30-100um)
- Sheet jets

Not so Standard:

- Thinner jets
- Drop on demand
- Solid targets

Resolution (Instrument Response):

Standard: 65 to 100 fs

Not so Standard: <65

Pump Laser:

Standard:

- Collinear (2 deg)
- Fundam&Harm 800,400,266nm
- OPA (480-2400 nm)

Not so Standard:

- Not collinear
- THz
- 8 ns laser (410-2200 nm)

Parameter Table for the XPP Standard Configuration

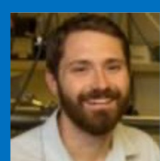
Sample	Sample(S) description	
	Temperature range [C]	
X-ray Parameters	X-ray Energy	Fixed to 9(Cu k-edge)-13 keV
	X-ray Pulse Duration	Fixed to ~50fs
	Mono bandwidth [meV] (default 600 meV with Diamond(111))	
	X-ray Focal spot size within 10 to 200 μm	
	X-ray polarization Vertical(default), horizontal or Circular (including switching capability)	
Detector	Detector positioning range, List of Bragg reflections and typical scattering angles.	
Optical beam parameters	Wavelength [nm]	
	Pulse duration [fs]	
	Maximum Pulse Energy [μJ]	
	Focal size (FWHM) [μm]	
	Polarization requirements?	
	Minimum fluence on sample [mJ/cm^2]	
	Geometry	Collinear or Non-collinear up to 7 degree
X-ray Beam Time	Number of shifts [1 shift = 12 hr]	
Any additional comments		

Parameter Table for the XCS Standard Configuration

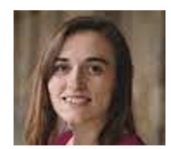
Sample & sample delivery	Sample(S) description	
	Round jet diameters [20, 30, 40, 50, 75, 100, 125, 150, 175, 200, 250, 300, 500] μm	
	Flat sheet jet thickness [19, 25, 38, 50, 75, 100, 125, 175, 188, 250] μm	
Scattering	[yes/no]	
	Maximum Q [$Å^{-1}$]	
X-ray Emission Spectroscopy	[yes/no]	
	Which Emission line(S) : Mn : $K\beta_{1,3}$, $K\beta_{2,5}$ and K; Fe : $K\beta_{1,3}$ and $K\beta_{2,5}$; Co : $K\beta_{1,3}$ and $K\beta_{2,5}$; Ni : $K\beta_{1,3}$ and $K\beta_{2,5}$; Ti : $K\beta_{1,3}$ and $K\beta_{2,5}$; V : K_α.	
X-ray Parameters	X-ray Energy	Fixed to 9.5keV
	X-ray Pulse Duration	Fixed to ~50fs
	Monochromatic or Pink	
	X-ray Focal spot size within 2 to 100 μm	
Optical beam parameters	Wavelength [nm]	
	Pulse duration [fs]	
	Maximum Pulse Energy [μJ]	
	Focal size (FWHM) [μm]	
	Polarization requirements?	
	Minimum fluence on sample [mJ/cm^2]	
	Geometry	Collinear
X-ray Beam Time	Number of shifts [1 shift = 12 hrs]	
Any additional comments		

Gas Phase Photochemistry in the SRD Department Structure

AMOS Department
(James Cryan)

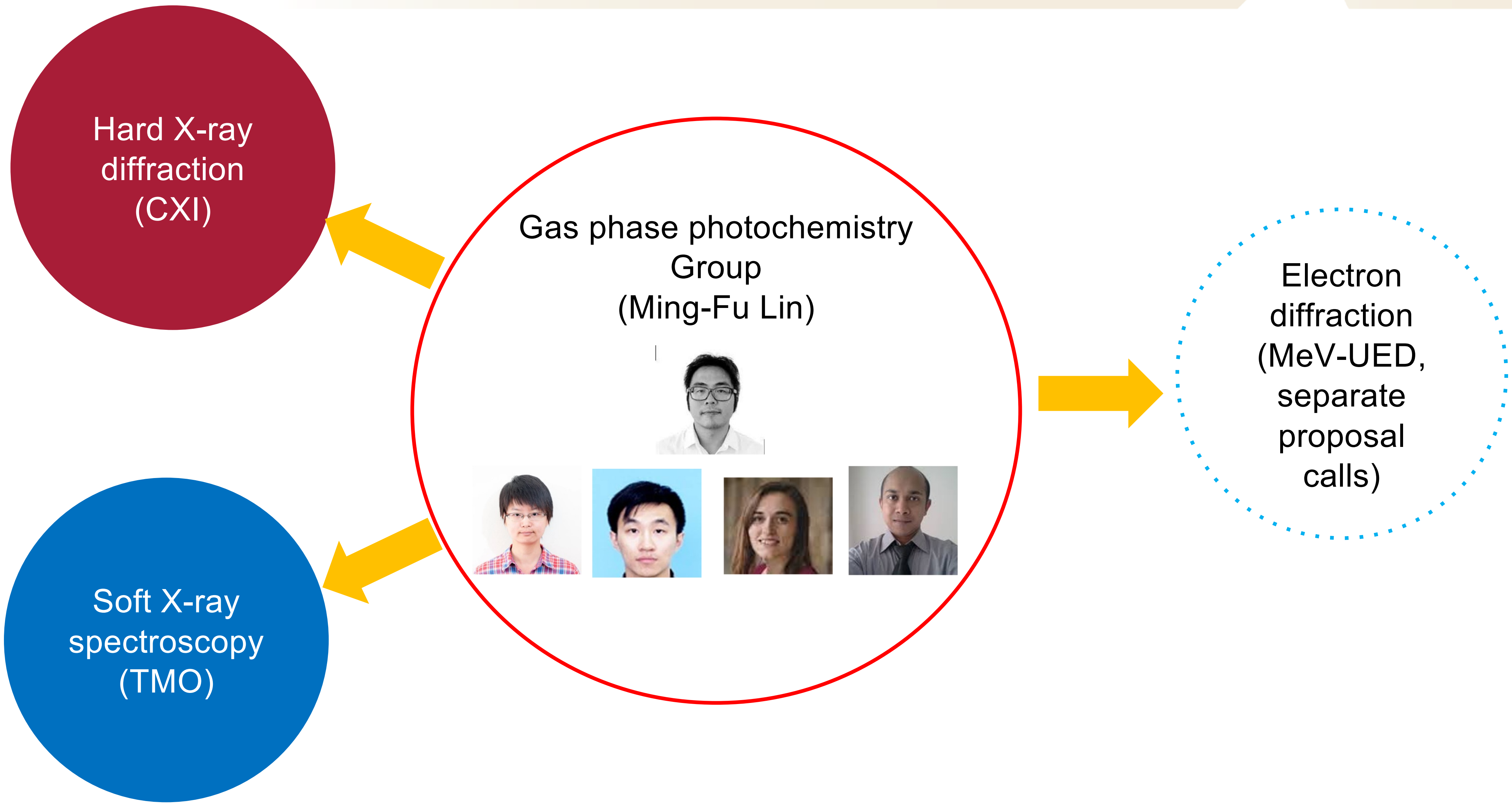


Gas phase photochemistry
Group
(Ming-Fu Lin)

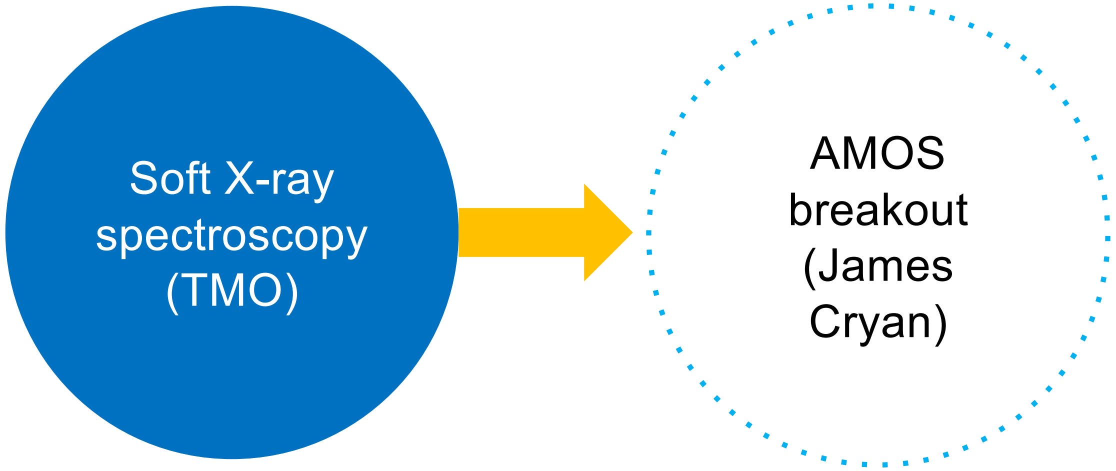


Chemical Sciences
Department
Thomas Wolf

Techniques and Instruments Supported by the Group



Soft X-ray Early Science Run 21



Gas Phase Chemistry at CXI

SLAC

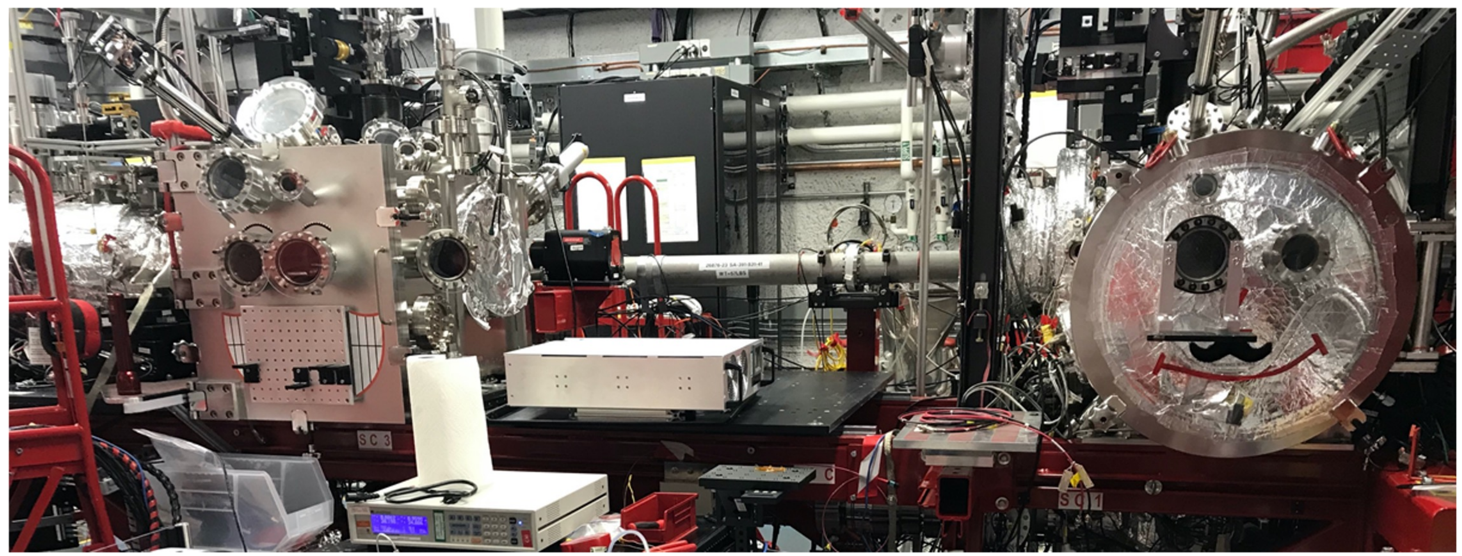
[CXI - Coherent X-ray Imaging | Linac Coherent Light Source (stanford.edu)]

Primary considerations:

- Low background scatter – Vacuum environment at hard X-ray energies with numerous slits for a clean focal spot
- Short Pulse UV capabilities

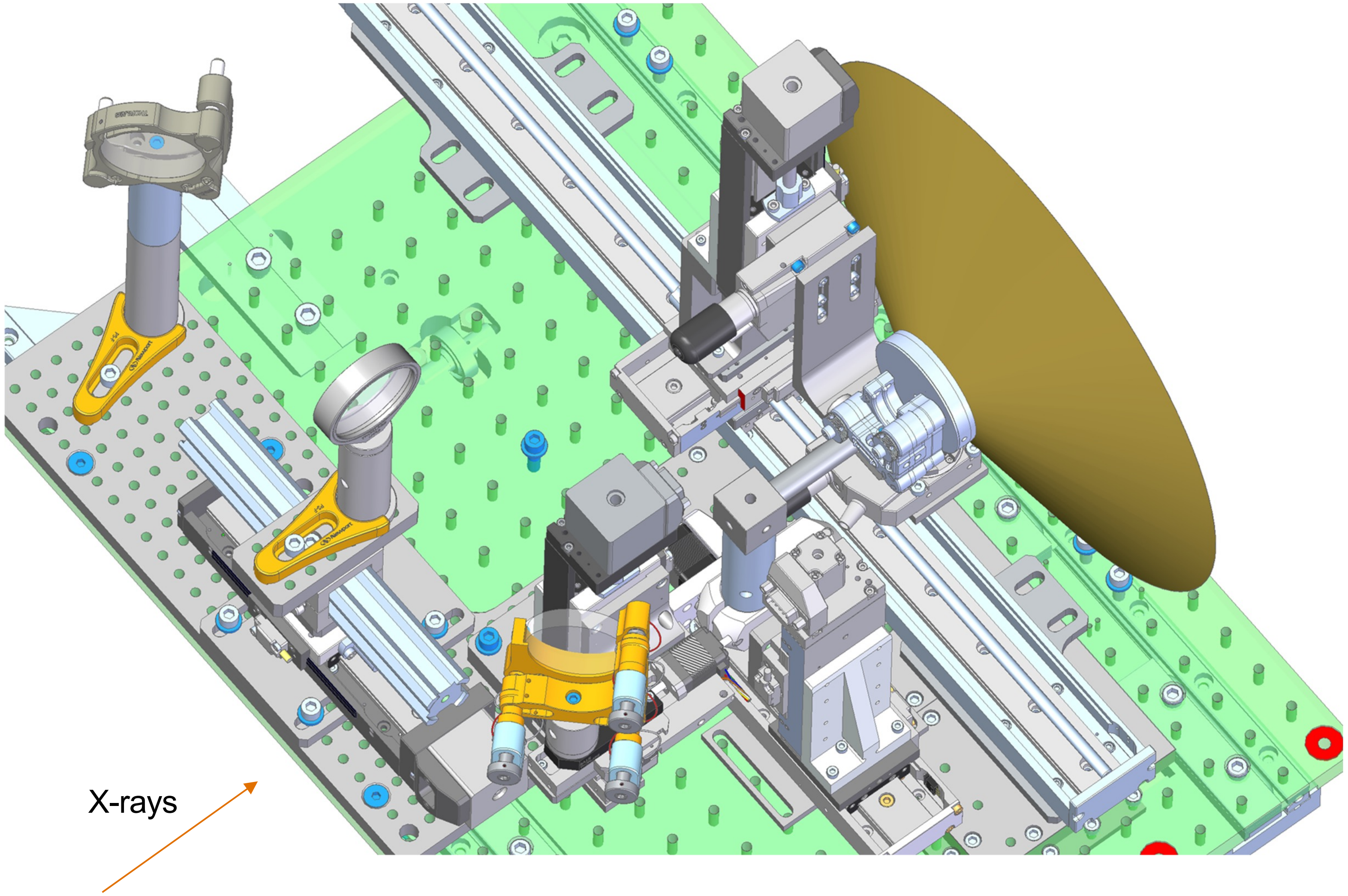
Standard configuration for gas phase chemistry:
[CXI Standard Configuration | Linac Coherent Light Source (stanford.edu)]

- Photon energy
  - 7keV-11keV (1 µm focal spot) – KB mirrors (reflective optics)
  - 11keV-25keV (2-3µm – 50µm focal spot) – CRLs

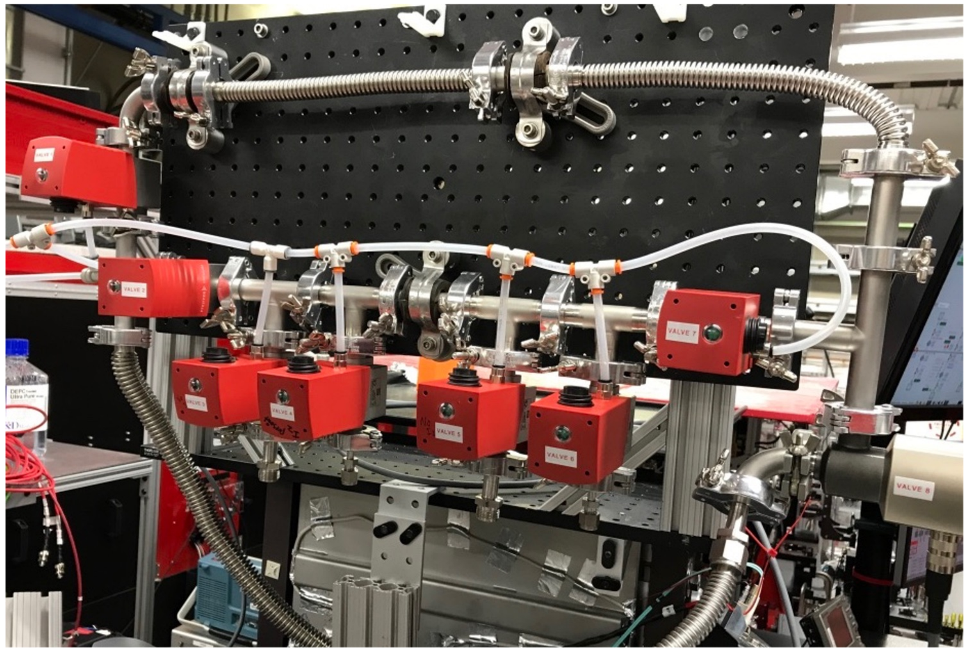


Standard Configuration

- Gas cell
- Be exit window downstream
- Pt pinhole entrance
- Additional Pt pinhole upstream
- Scattering cone
- UV pump propagates in-line with the X-rays
- Fully controllable sample delivery manifold

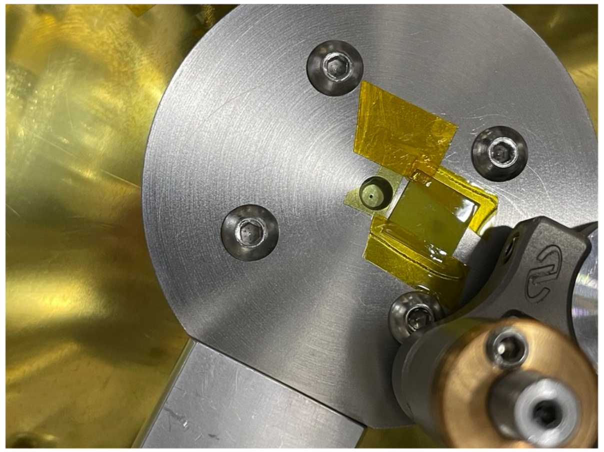


Standard Configuration



gas manifold - accommodates 4 samples

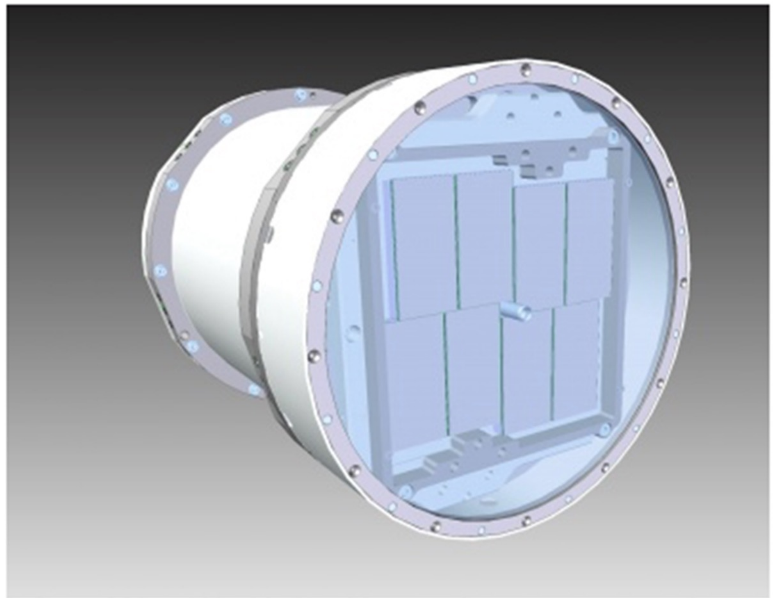
Gas cell, pinhole, scattering cone



gas cell, entrance pinhole and frosted YAG for spatial overlap

Standard Configuration

- Detector – 4M Jungfrau detector
  Jungfrau | Linac Coherent Light Source (stanford.edu)
  - Adaptive gain
  - background is <1 photon / image with proper alignment
- in-line X-ray spectrometer available as needed
- Downstream I0 monitor



New for Run 21

- Prefocusing lenses in the XRT to increase flux when using the CRLs by avoiding losses due to the clear aperture of the CXI CRLs
- Downstream monitor of the UV pump power (after sample), likely in SSC (downstream chamber)

Short Pulse UV development

Short Pulse UV capabilities are under constant development

CXI Specifications | Linac Coherent Light Source (stanford.edu)

Phase 1: Improving the time resolution of the 3rd and 4th harmonics

	Current Pulse Width (FWHM)	Expected Performance (FWHM)
267 nm (3ω)	~80 fs	~35 fs
200 nm (4ω)	~120 fs	~50 fs

Phase 2: Generating tunable deep UV pulses

	Current Capability	Target Capability
245-260 nm	Available Run 21	~35 fs
220-245 nm	Possible for Run 21*	~40 fs
280-330 nm	Possible for Run 21*	~35 fs

Please contact CXI team member about your UV laser needs!

Job Opening for an Associate Scientist

- Develop gas phase photochemistry research and instrumentation
- Conduct and support experiments at UED, CXI, and TMO
- Join the UED instrument team

https://erp-hprdext.erp.slac.stanford.edu/psp/hprdext/EMPLOYEE/HRMS/c/HRS_HRAM_FL.HRS_CG_SEARCH_FL.GBL?Page=HRS_APP_JBPST_FL&Action=U&FOCUS=Applicant&SiteId=1&JobOpeningId=4908&PostingSeq=1

Contact:

- Thomas Wolf (thomas.wolf@slac.stanford.edu)
- James Cryan (jcryan@slac.stanford.edu)